

# New excitations in bcc $^4\text{He}$ - an inelastic neutron scattering study

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We report neutron scattering measurements on bcc solid  $^4\text{He}$ . We studied the phonon branches and the recently discovered "optic-like" branch along the main crystalline directions. In addition, we discovered another, dispersionless "optic-like" branch at an energy around 1 meV ( $\sim 11\text{K}$ ). The properties of the two "optic-like" branches seem different. Since one expects only 3 acoustic phonon branches in a monoatomic cubic crystal, these new branches must represent different type of excitations. One possible interpretation involves localized excitations unique to a quantum solid.

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Elementary excitations of quantum solids show unique properties arising from the large zero point vibration of the atoms. The interatomic potential is highly anharmonic, and the atoms feel strong short range correlations, due to the repulsive part of the potential[1–3]. The "self consistent phonon" theory developed to treat this problem yielded phonon dispersion curves in a reasonable agreement with experimental data available at the time[4, 5]. Recently, an additional, "optic-like" excitation branch was discovered[6] along the [110] direction of bcc  $^4\text{He}$ . This is puzzling since on general grounds, only *acoustic* phonon branches should exist in a mono-atomic cubic crystal. Several interpretations of the new excitation were discussed[6] in terms of multiphonon effects[3], or localized excitations unique to a quantum solid. The latter include an isotropic vacancy band[7–9] or anisotropic local modes in the [110] direction associated with correlated zero point motion[10]. An analysis of the data showed that multiphonon effects were not the source of this new branch[6]. In addition, each of the models involving localized excitations was consistent only with some facet of the data[6]. Localized excitations in solid He are of particular interest in view of the recent reports of the "supersolid" phase[11]. Our motivation to do additional experiments was twofold: first, the new "optic-like" branch was measured only along the [110] crystalline direction. The anisotropy of this new branch can be determined only if it is investigated in all directions. Second, since the new branch was largely unexpected, perhaps other such features may be found.

In that spirit, we performed inelastic neutron scattering experiments on bcc  $^4\text{He}$  using the IN-12 and IN-14 triple axis spectrometers at the ILL. Single crystals of high purity  $^4\text{He}$  (less than 0.1 ppm  $^3\text{He}$ ) were grown and oriented in the beam. The growth orientation of these crystals was a priori random. In most cases, the

limitations of orienting the crystals by tilting the cryostat allowed us to measure only along the [110] direction. In order to access other crystallographic directions, we added a home built cold goniometer to the sample stage. Two versions of the apparatus were built, the first one allowing an additional  $\pm 12^\circ$  tilt of the cell in an arbitrary direction. This apparatus was used for inelastic scattering along the [110] direction carried out on the IN-14 spectrometer. The second version, allowing an additional  $\pm 45^\circ$  tilt along one axis, was used for measurements along [100] and [111] done in a separate experiment on the IN-12 spectrometer. Crystals of low density  $^4\text{He}$  ( $21\text{ cm}^3$  molar volume), several  $\text{cm}^3$  in size were grown from the superfluid at  $T=1.640\text{ K}$ , where the temperature width of the bcc phase is maximal ( $\sim 50\text{ mK}$ ). In order to get a high quality single crystal, the solid was further annealed overnight on the melting curve. The final crystal was composed of two large grains misaligned by  $20^\circ$ , with volumes of about 2 and  $4\text{ cm}^3$ . The FWHM of the rocking curve was about  $30'$  and  $40'$  for the two grains. Scattering experiments were conducted at the same temperature. In addition to the crystal, the cell contained a small amount of superfluid helium (less than 1% of the volume) in order to reduce any temperature gradients across the sample. In particular, we were able to get very high resolution data along [001] and [111] from the *same* crystal, which could be oriented to access either the {002} scattering plane or the  $\{1\bar{1}2\}$ . The energy of the incident beam could be varied in the range of 2.3 to 14 meV. Constant-Q scans were done at a fixed momentum of the scattered neutrons ( $k_F$ ). In the high resolution scans we used a cooled Be filter to remove  $\lambda/2$  contamination. Focusing techniques were used to enhance the scattering intensity. The highest instrumental resolution was 0.1 meV (FWHM) with  $k_F=1.2\text{ \AA}^{-1}$ . In order to ascertain that the new features described in the following are not spurious, we carried out extensive background measurements by repeating the inelastic scans in the various directions once with an empty cell, and once with the cell full of liquid. We found that the new features disappeared once the crystal was molten, hence they're not connected with scattering from the liquid or from the

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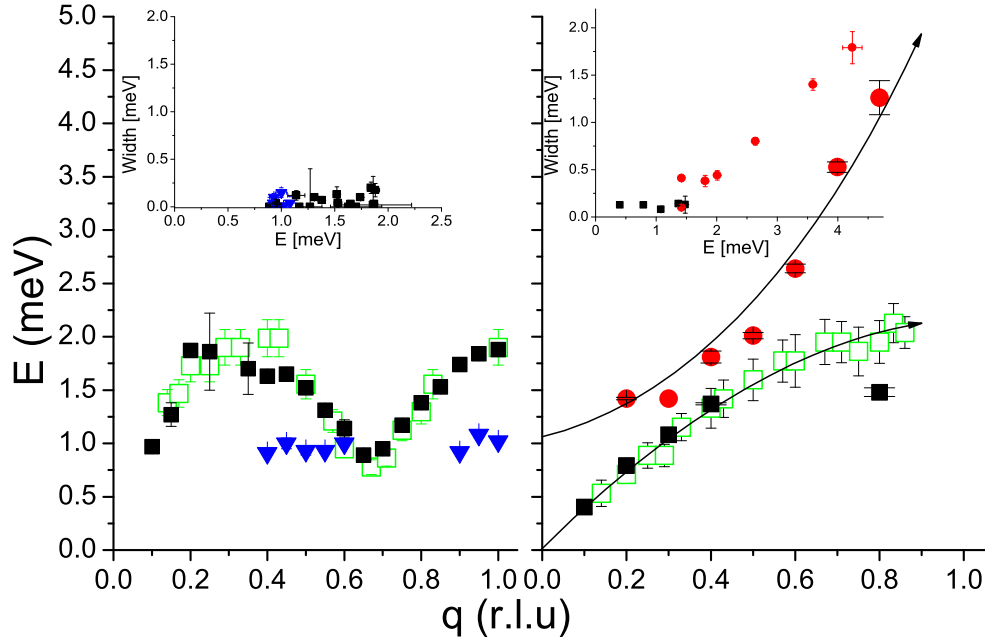


FIG. 1: (color online) Dispersions curves along the [111] direction. Left panel: longitudinal polarization. Right panel: transverse polarization. In both panels, solid black squares are data points for the phonon branches, and the open green squares are results of PIMC simulations of the dispersion curves[14]. The inverted blue triangles in the left panel are data points for the LOB. In the right panel, the red circles are data points for the HOB. The solid lines are a guide to the eye. The inset shows the energy linewidth of the features seen in the inelastic spectra. Note that broadening occurs only for the polarization where the HOB is observed(right panel).

walls of the cell. Bragg scattering measurements of the crystals failed to show any traces of the hcp phase. We conclude therefore that the new features are a property of the bcc solid.

Our measurements of the phonons fill some gaps in the available data for the phonon dispersion curves[4, 5, 12]. These results will be described in detail elsewhere[13]. In this paper, we focus on the new optic-like branches. In order to assign the various peaks seen in the neutron scattering data, we performed Path Integral Monte Carlo (PIMC) simulations[14] of bcc  $^4\text{He}$  to determine the contribution of single phonons to the dynamic structure factor  $S(q, \omega)$ . Figure 1 shows the dispersion relations of the L and T phonons along [111], along with the two "optic-like" branches observed in these experiment. The left panel shows the L branch and another, dispersionless branch at an energy around 1 meV ( $\sim 11\text{K}$ ). We label this new branch as the "lower optic-like branch" (LOB). Regarding the phonons, the data for the L branch is in excellent agreement with the simulations. The measured energy linewidth of the phonon peaks in the spectra (see inset) is small, limited by the experimental resolution. Turning to the right panel of Figure 1, again two branches are seen, the T[111] phonon branch and another, "optic-like" branch which we identify with the branch observed previously[6] along [110]. We label this branch as the "higher optic-like branch" (HOB). It is seen that the data for the T branch agrees with the simulations only for energies less than the minimum energy of the HOB branch.

Similarly, the inset shows that the linewidth increases quite strongly above this minimum energy. Quite obviously, one cannot assign the points on the dispersion curve with a large linewidth to single phonons. The increase of the linewidth was observed also for the L[100] branch where the HOB excitation was also seen.

First, we discuss the HOB "optic-like" excitation branch, previously observed only along the [110] direction[6]. In the present work, this branch was observed in scans measuring the T(111) and L(100) phonons, while in the scans for L(111) and T(100) it was absent. Hence, this excitation is anisotropic. The dispersion of this branch, when plotted together with the usual phonons is suggestive of mode coupling. This is evident for example in the right panel of Figure 1 showing the T[111] branch, as well as in earlier data[6] along [110]. Additional support for this suggestion comes from the results plotted in Figure 2 showing the polarization dependence of the HOB along [110]. It is seen that in scans with the T1 polarization, this branch (labelled HOB-T1) shows little dispersion, while in scans with a longitudinal polarization (labelled HOB-L) the dispersion is significant. Since the maximal energy of the T1[110] branch is 0.6 meV ( $\sim 7\text{K}$ ), while the minimal energy of the HOB is 1.23 meV ( $\sim 14\text{K}$ ), the T1 and HOB branches do not cross. Hence, mode coupling with the T1 branch should be weak. On the other hand, the HOB and the L(110) branch do cross and so the coupling with the L(110) should be stronger. Both the broadening shown in the

right panel of Figure 1 and the data in Figure 2 are consistent with this idea.

In order to determine the intrinsic dispersion of this branch, we attempted to simultaneously fit the dispersion relations of the HOB excitation and the phonon branches using a mode coupling approach. We took the coupling of the branches to be in the form  $\Delta/((\omega_{HOB}^2 - \omega_{ph}^2) + iD)$ , where  $\Delta$  is the coupling constant,  $\omega_{HOB}$  and  $\omega_{ph}$  denote the energies of the HOB and phonon branches respectively, and  $D$  is connected to the damping which prevents a singularity at mode crossing. This particular form is not based on a specific model of this excitation, hence it is useful only to gauge the relative strength of the coupling between the various branches. After trying several similar forms[15, 16], two conclusions can be drawn; first, it was possible to fit the coupled branches only by assuming that the HOB has a finite dispersion. Second, the value of the coupling constant  $\Delta$  is different for different phonon branches. For the [110] direction, it is about twice as big as for the other directions. The ratio  $\Delta/D$  however, is approximately constant for all the branches. Consequently, we believe that the intrinsic dispersion of the HOB is that observed with the T1(110) polarization (Fig. 2). The dispersion is weak, and within our resolution can be fitted either to a linear or quadratic dependence on  $q$ . The linear fit,  $E(q) = \epsilon_0 + v_G q$ , yields a group velocity  $v_G \sim 68$  m/sec. The quadratic fit,  $E(q) = \epsilon_0 + \hbar^2 q^2 / 2m^*$ , appears marginally better, with  $\epsilon_0 = 1.20$  meV and  $m^* \simeq 0.7 m_4$ . The total bandwidth is about 0.4 meV ( $\sim 4.6$ K). To conclude, the HOB branch is anisotropic, in the sense that is observed only in certain directions, and seems to couple more strongly to phonons along [110]. This branch has a finite intrinsic dispersion.

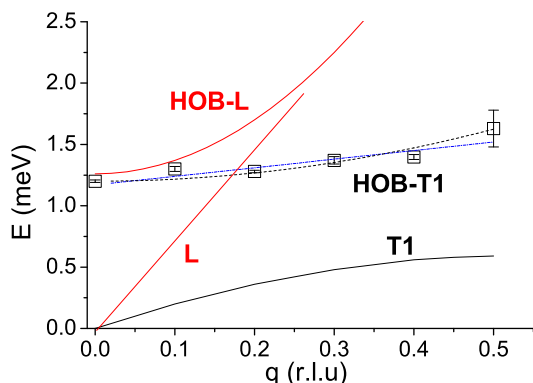


FIG. 2: (color online) Dispersion curves of the Higher Optical Branch (HOB) along [110] with two polarizations. The lines labelled HOB-L and L are smoothed values of the HOB and the L(110) phonon branches from Reference [6]. HOB-T1 denotes data points from this work for the HOB with T1 polarization, while the line labelled T1 represents smoothed values of the T1(110) phonon branch. The dashed black line is a linear fit to the HOB-T1 data, and the dash-dot blue line is a quadratic fit.

We now discuss the second new feature, namely the

LOB (see left panel, Figure 1). This excitation branch appears dispersionless within our resolution, with an energy of  $0.95 \pm 0.1$  meV [ $\sim 11$ K]. Typical scans showing the LOB are plotted in figure 3. This new excitation was seen in scans measuring the T(100), L(100) and the L[111] phonons while being absent in scans along the [110] direction, even at the highest resolution[6]. Hence, it is also anisotropic in the same sense as the HOB branch. In this context, we also tried to examine the possibility that this mode is excited only when the incident beam is along some specific direction. We found that in the {002} scattering plane, the excitation was observed when the incident beam was around the (110) direction, while in the {112} plane the incident beam was around the (021) direction. These directions are not equivalent, so the excitation of the mode does not seem to be linked to the direction of the incident neutrons. Another remark is that near the minimum of the dispersion curve of L[111] ( $q=2/3$  r.l.u.), there is an inherent mixing of the L[111] phonons with phonons originating in the {112} scattering planes. Hence, these planes may contribute to the intensity of the L[111] phonons and perhaps add another feature to the scans. However, the LOB is seen also in the [100] direction, where there is no phonon mixing. Additional discussion of these points will be presented elsewhere[13]. The energy linewidth of this new feature is small, limited by the instrumental resolution. The intensity of the LOB decreases with reduced resolution, and at low resolution it was not observed. This may perhaps explain why it was not seen in the past[4]. It may also be the reason why the LOB was not seen in the scans measuring the T[111] branch, where we were not able to work with a high resolution. The fact that the LOB is dispersionless and its linewidth remains small even when it crosses a phonon branch implies that there is no interaction between the LOB and phonons. In that respect, the LOB and the HOB are different types of excitations.

The absence of dispersion of the LOB suggests that this excitation is localized. Localized excitations can be point defects or more complex entities, e.g. "local modes"[17]. In usual materials, phonon energies are in the meV range while those of point defects are in the eV range. Consequently, in usual materials point defects cannot be observed in scans measuring phonon branches. In solid He, these two energy scales are very similar, so in principle point defects could be excited by cold neutrons. The energy of the LOB is indeed similar to the typical energy of a point defect in bcc  $^4\text{He}$  measured by different techniques[18, 19]. A neutron incident on the solid can create a vacancy by knocking a single atom away from its lattice site. However, single particle excitations have a large energy width because the final state of the atom is in the continuum[4]. Hence, this possibility is not consistent with the small linewidth of the LOB. The same argument applies to creation of vacancy-interstitial pairs (Frenkel pairs). The formation energy of a Frenkel pair depends on the interstitial-vacancy distance, which can take different values. Thus, one expects a broad feature

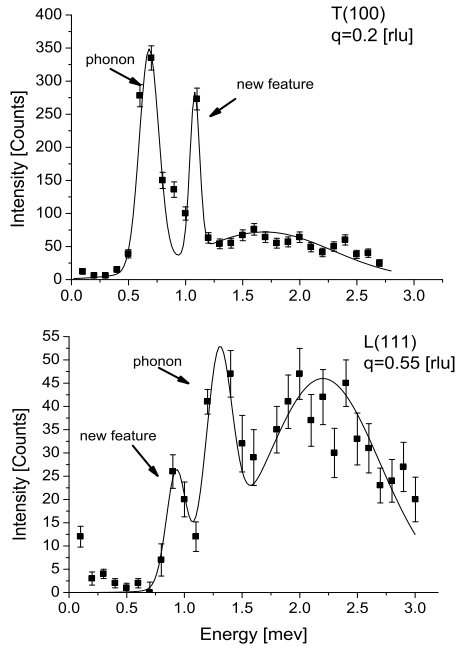


FIG. 3: Neutron groups in the [100] and [111] directions showing the new low energy "optic like" (LOB) excitation. The squares are data points and the solid line is a gaussian fit. The "new feature" marks the LOB. The instrumental resolution in the upper panel is higher.

in the scattering intensity vs. energy rather than the narrow peak which is observed (Fig. 3). In addition, recent simulations of point defects in bcc He indicate that a vacancy branch has a considerable dispersion[20]. This result supports our claim that the dispersionless LOB branch is probably not associated with creation of va-

cancies. Another possibility is that the neutrons excite some resonant mode[17, 21], namely internal vibrations of split interstitials or vacancies already present in the crystal. Formation energies and resonant mode energies of point defects should be similar, and in addition, the energy width of resonant modes is small. Some qualitative evidence of the presence of such excitations was observed in our simulations of crystals containing a large number of interstitials[13]. Hence, although we did not find any analytical calculations of these modes for solid He, this possibility is of interest. Finally, a feature similar to the LOB may have been observed in neutron scattering from the hcp solid phase at a temperature of 100 mK[22]. Resonant modes, if they exist, should be observed in both hcp and bcc He. Further investigation of these features in both solid phases may help to understand their origin.

In conclusion, we investigated the recently discovered "optic-like" excitation branch(HOB) and found it to be anisotropic and weakly dispersive. In addition, another new excitation branch was discovered (LOB). This branch is also anisotropic, dispersionless, and with a very small linewidth. In contrast with the HOB, the LOB does not couple to phonons. One possible interpretation is that these branches are associated with point defects or excitations thereof. However, none of the existing models is detailed enough to allow meaningful comparison with the data.

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